

Fabrication of stable and reproducible sub-micron tunnel junctions

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We have performed a detailed study of the time stability and reproducibility of sub-micron $Al/AlO_x/Al$ tunnel junctions, fabricated using standard double angle shadow evaporations. We have found that by aggressively cleaning the substrate before the evaporations, thus preventing any contamination of the junction, we obtained perfectly stable oxide barriers. We also present measurements on large ensembles of junctions which prove the reproducibility of the fabrication process. The measured tunnel resistance variance in large ensembles of identically fabricated junctions is in the range of only a few percents. Finally, we have studied the effect of different thermal treatments on the junction barrier. This is especially important for multiple step fabrication processes which imply annealing the junction.

Sub-micron sized metal/ AlO_x /metal tunnel junctions are used in a wide range of applications in many fields, from superconducting and single charge electronics to calorimetry, nanomagnetism and spintronics. However, the time instability of the AlO_x barrier is a frequently reported drawback. Systematic measurements of junction aging showed that the resistance of the tunnel barrier could double its value within a period of days¹. Similarly, a reduction of the capacitance C of the junctions was also observed². The junction aging is usually associated with two types of phenomena. Either (i) the diffusion of oxygen atoms from the oxide barrier to the electrodes³ or (ii) the change of the chemical composition of the barrier, by absorption or desorption of atoms or molecules other than oxygen⁴. It has been shown that mechanism (i) plays a secondary role in the aging of the barrier, only accounting for the slow drift of the junction parameters at long time scales. Moreover, this slow diffusion of oxygen atoms can be suppressed by surface nitridation of the electrodes⁵. Mechanism (ii) is believed to play the dominant role in the aging of the junction barrier. Vacuum anneals at temperatures between 200 °C and 450 °C will accelerate the relaxation processes in the AlO_x barrier and the resulting junctions show improved characteristics^{1,6}.

The origin of the junction contamination, which leads to the chemical relaxation in mechanism (ii), has been up to now subject of speculation. Using a standard bilayer double angle evaporation technique⁷, Koppinen et al.¹ observed a decrease in the junction aging when the substrate was cleaned with an oxygen plasma just before the electrodes evaporation. This observation suggests that the aging of the barrier is linked to the existence of resist residues in the vicinity of the junction during the deposition. Indeed, by using fabrication methods which avoid the direct contact between the photoresist and the insulating layer, it is possible to obtain perfectly stable junctions^{8,9}. In the following we will show that by aggressively cleaning the substrate before the Al deposition, we could obtain perfectly stable $Al/AlO_x/Al$ junctions, using the standard bilayer double angle evaporation technique. Further on, we will show evidence that the junction aging is a consequence of the oxide barrier contamination from resist residues.

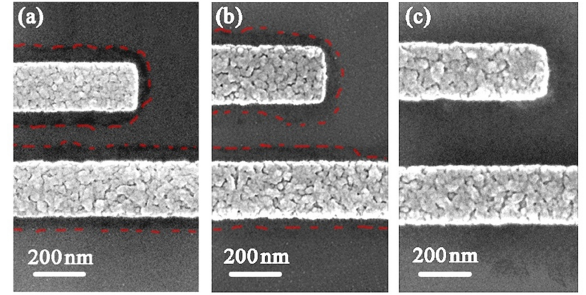


Figure 1: SEM images of zero angle deposited Al wires, for three different development sequences. The images are taken at high contrast in order to observe the resist residues, which are visible as black traces. For clarity, we highlighted them with the red dotted line. (a) Standard MIBK/IPA 1/3 (30s) and IPA (60s) development. (b) After the standard development we added an Ethanol/IPA 1/3 (60s) step. We notice that the wafer is significantly cleaner, even though some resist traces can still be seen. (c) After the standard development we performed an oxygen plasma RIE. Notice that the surface of the wafer is now completely resist free.

We fabricate the samples using the standard *Dolan bridge* technique⁷. We produce the suspended resist mask using a PMMA/PMMA-MAA bilayer, deposited on a Si/SiO_2 substrate. The circuit patterns are written using e-beam lithography. After development we perform two shadow evaporations of Al in a UHV system with a base pressure of 10^{-9} mBar. In order to obtain tunnel junctions, between the two evaporations we oxidize the first Al layer in a pure oxygen atmosphere at different pressures $P_{oxydation}$, depending on the sample (see Table I).

In Fig. 1a we show a SEM image of Al wires deposited after a standard development of the resist bilayer. In the vicinity of the electrodes we see a significant contamination with residual resist. In order to clean these resist residues, we have tried two processing steps. First, after the standard development, we plunge the wafer for 1 minute in a mixture of Ethanol and IPA. Ethanol is an aggressive solvent and it removes large volumes of resist from the undercut. In Fig. 1b we show a SEM image of the resulting Al wires. We notice that we have

removed part of the residues, but the substrate is not yet completely clean. The second wafer cleaning step that we tried is the Reactive Ion Etching (RIE) in oxygen plasma. The plasma parameters were adapted to ensure a slow, isotropic etching¹⁰. This is not obvious because we have to clean a relatively large undercut surface, compared to the small access opening in the top resist. The SEM image of the resulting Al wires, presented in Fig. 1c, shows that the oxygen plasma RIE has completely cleaned the resist residues.

Using the oxygen plasma RIE cleaning step, we have fabricated several sets of ~ 100 junctions that we have contacted and measured individually at room temperature. The geometry of the sample (Fig. 2) was opti-

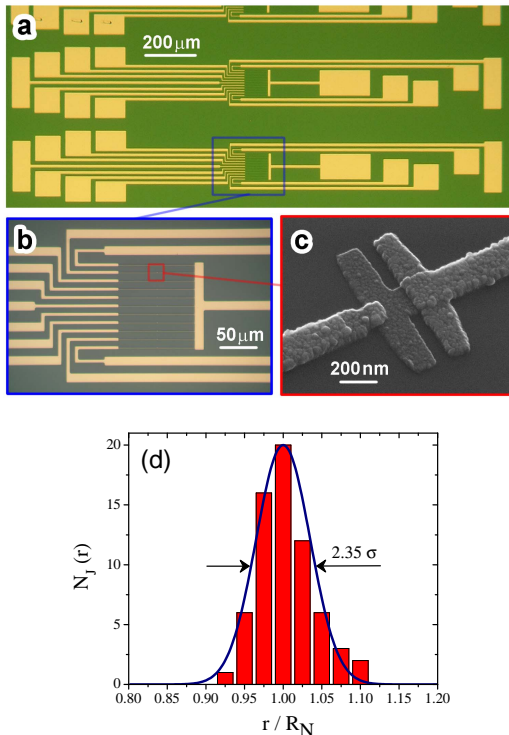


Figure 2: Images of the circuit fabricated to measure the time stability and the variance of the junction's tunnel barrier. In (a) and (b) we show optical images of one of the junction sets at different magnifications. (c) SEM image of one junction. (d) Measured histogram of the tunnel barrier resistance, for identically fabricated junctions with a surface of $0.03 \mu\text{m}^2$. We also represented the width at half height for the Gaussian fit (blue line), which is $\sim 2.35 \cdot \sigma$, where σ is the variance. For clarity, the x -axis is reported in units of the mean value for the resistance: R_N .

mized to enable the packing of a large number of identical junctions in a small area of the wafer, in order to minimize the variations of resist thickness. The surface S of the junctions depends on the sample, ranging from $0.02 \mu\text{m}^2$ to $0.2 \mu\text{m}^2$. At low temperatures, the junctions have critical current densities in the range of $\sim 1 \mu\text{A}/\mu\text{m}^2$. This class of $\text{Al}/\text{AlO}_x/\text{Al}$ junctions is widely used in superconducting quantum electronics¹¹.

In Fig. 2d we present a typical histogram for the tunnel resistance r measured at room temperature. The mean value of the resistance $\langle r \rangle = R_N$, depending on the sample, is of the order of a few $\text{k}\Omega$. The distribution is well fitted (see the blue line in Fig. 2d) by a Gaussian law: $N_J(r) = N_{\text{tot}} \left(1/\sqrt{2\pi\sigma^2} \right) \exp \left[-(r/R_N - 1)^2 / 2\sigma^2 \right]$. Here σ^2 is the variance, in units of R_N^2 and N_{tot} is the total number of measured junctions. The measurements in Fig. 2d can be well fitted by taking the value $\sigma = 3.5\%$. All measured junction ensembles showed similar variance values, within a few percents.

We measured the average tunnel resistance of the junctions at room temperature, directly after the fabrication process $R_N(0)$ and during the following 4 weeks $R_N(t)$. We studied several ensembles of ~ 100 junctions (see Fig. 2) for which the fabrication conditions and the storage were identical. The results of our observations are presented in Table I. For the junctions

chip	$P_{\text{oxydation}}$	storage	$R_N(0) \times S$ [$\Omega \times \mu\text{m}^2$]	$\frac{R_N(t) - R_N(0)}{R_N(0)}$
A	$5 \cdot 10^{-2} \text{ mbar}$	vacuum	210	0%
B	$4 \cdot 10^{-2} \text{ mbar}$	atm	180	0%
C	$5 \cdot 10^{-3} \text{ mbar}$	atm	160	+10%

Table I: Measurement of junction stability, during 4 weeks after fabrication. The error bar for the measurement is $\sim 1\%$. The oxidation time for all samples is 5 minutes.

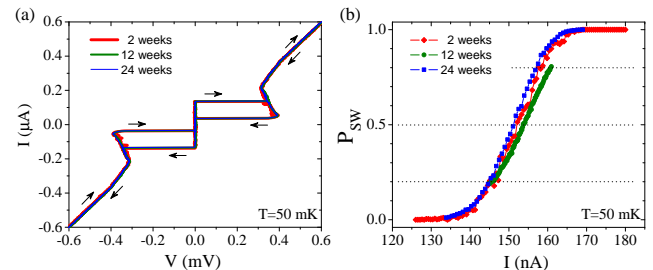


Figure 3: Current-voltage curves (in a) and switching current measurements (in b) for the same junction from group A, measured 2 weeks (in red), 12 weeks (in green) and 24 weeks (in blue) after the fabrication. Between the measurements, the junction was stored at room temperature and atmospheric pressure.

of chip A and B we did not observe any aging. For sample C we measured a slight increase in the tunnel barrier, which we attribute to the adsorption of oxygen from the surrounding atmosphere. This adsorption is favored by the high reactivity of the barrier, which was very weakly oxidized. All the measured tunnel barriers are practically stable if compared to the previously reported increases of 100% up to 300% within the first weeks after the fabrication^{1,2}.

One of the junctions from group A was measured at $T = 50 \text{ mK}$ in the superconducting regime, 2 weeks, 12

weeks and 24 weeks after the fabrication. The measured *IV* characteristics and the switching curves are plotted in Fig. 3. Notice that the results of the 3 measurements at $T = 50$ mK are practically identical. We did not observe any aging of the tunnel barrier, the critical current or the retrapping current. The switching current and the width of the switching curves, both depend on the junction capacitance C , and show no aging between the three measurements. Thus, we conclude that the capacitance of the junction is also completely stabilized. From the small difference of ± 5 nA between the three switching curves, we infer the error bar of our measurement, which is smaller than 5%.

In the following, we study the effects of annealing on the oxide tunnel barrier. We have measured the effect of vacuum thermal cycles on the tunnel barrier of our junctions. The steps of one thermal cycle are the following: 10 minutes heating, 10 minutes regulating the temperature to a constant value and 20 minutes cooling down. In Ref.¹ it has been shown that the resistance of samples annealed at 200 °C and 400 °C increased respectively by 50% and 300%. However, only the latter were completely stable in time. We report fundamentally different results for junctions fabricated using the RIE cleaning. For sample *B*, after annealing at 200 °C we observe no change in the tunnel barrier resistance. This confirms the stability of our junctions. After annealing at 400 °C we observe a decrease of the barrier by almost 50%. This decrease is expected as the oxygen diffusion in the barrier starts to be thermally activated for $T \gtrsim 300$ °C^{4,12}.

We have annealed at 200 °C the junctions of a forth sample, *D*, similar to *C*, which was covered by a *PMMA* resist layer. After the anneal, we observed a significant decrease of 26 % of the tunnel resistance. This could be explained by the contamination of the

junction with hydrates ($-OH$) from the resist, which combine with the oxide to form hydroxides (for example $Al_2O_3(H_2O)$ or $Al_2O_3 \cdot 3(H_2O)$) and thus decrease the barrier height, as suggested in Ref.⁴. These junctions were unstable and we measured a slow relaxation of the barrier value towards the initial state. In one week, the barrier increased by 3%. After re-annealing the sample *D* under a *PMMA* resist layer at 200 °C, the value of the barrier returns to lower values. After 4 weeks, the barrier relaxes again to a value 6% larger. This reproducible increase-relaxation process for the barrier value, after each anneal under a *PMMA* resist layer, can be explained as follows. During the annealing, the junction absorbs hydrates which account for the barrier decrease. After the lift-off of the resist, the slow desorption of hydrates during storage accounts for the measured relaxation of the barrier.

In conclusion, we have fabricated completely stable $Al/AlO_x/Al$ Josephson junctions, which were monitored for as long as 6 months, without observing any changes in the barrier parameters. The crucial processing step, allowing the fabrication of stable junctions, is the optimized, isotropic RIE oxygen-plasma cleaning of the resist residues, before the aluminum evaporation. We show that the junctions are stable to annealing up to 200 °C in vacuum. When annealed under a *PMMA* resist layer, the junction barrier decreases significantly and it also becomes unstable. Finally, measurements on large ensembles of junctions at room temperature show that the variance of the tunnel barrier height is in the range of only a few percents. This spread is considered acceptable for most types of superconducting circuits.

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